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**Step Flow and Interface Formation during the Growth of Heterojunction Nanowires**

**Frances M. Ross**

IBM T. J. Watson Research Center, Yorktown Heights, NY 10598, USA

Controlled fabrication of semiconductor nanowires that contain heterojunctions is the key to exciting electronic applications that make use of quantum dots and barriers. *In situ* transmission electron microscopy (TEM) provides a unique window into observing, and perhaps controlling, the formation of these complex nanostructures. We have therefore used *in situ* TEM to observe the growth of Si and Ge nanowires, and Si and Ge on III-V nanowires, through the vapor-liquid-solid and vapor-solid-solid mechanisms. Movies recorded during growth allow us to quantify wire nucleation, determine catalyst stability, measure nanowire surface structure and growth kinetics, and see the fascinating dynamic nature of the catalyst during heterostructure formation. We find that certain solid catalysts can be used to form compositionally abrupt Si/Ge and Si/SiGe interfaces, and we quantify the atomic-level step flow processes that take place during interface formation. We compare Si/Ge interfaces with group IV/III-V interfaces and discuss the implications of interface control for improved Si nanowire-based electronic devices.